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**TITLE:** Treating radioactive waste boric acid liq. by concentrating and mixing with phosphoric acid and its alkali metal (salt), and heating to form phosphate glass

**PATENT-ASSIGNEE:** MITSUBISHI ATOMIC POWER IND  
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**PATENT-FAMILY:**

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ABSTRACTED-PUB-NO: JP 63015200 A

**BASIC-ABSTRACT:**

Radioactive waste boric acid liq. discharged from atomic power stations, etc., is treated by a method where the waste liq. is concentrated and mixed with phosphoric acid and an alkali metal or an alkali metal salt of phosphoric acid, in amt. corresp. to amt. of boric acid present, so as to obtain a compsn., represented by region surrounded by line showing 20 mole% and 40 mole% B<sub>2</sub>O<sub>3</sub> and the line showing 20 mole% and 40 mole% X<sub>2</sub>O (where X is an alkali metal) in the glass composition range as represented by the Fig. I. The mixt. is heated to remove water and water of crystallisation, to form calcined body composed primarily of boron phosphate, followed by heating the calcined body to temps. above 600 deg.C to form phosphate glass.

USE/ADVANTAGE - Method can effectively and simply treat radioactive waste boronic acid liq. from atomic power stations, etc., by forming highly tough and stable solids at low cost, with emission of less radioactive fume and dust.

**TITLE-TERMS:**

TREAT RADIOACTIVE WASTE  
BORIC ACID LIQUID  
CONCENTRATE MIX PHOSPHORIC  
ALKALI METAL SALT HEAT FORM  
PHOSPHATE GLASS

**ADDL-INDEXING-TERMS:** SALT

DERWENT-CLASS: K07

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- 54. Invention Title: Phosphate Glassification Treatment Method for Radioactive Boric Acid Liquid Waste
  - 21. Application No.: Sho 61-157955
  - 22. Application Date: July 7, 1986
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## SPECIFICATION

- 1. TITLE OF INVENTION  
Phosphate Glassification Treatment Method for Radioactive Boric Acid Liquid Waste

- 2. WHAT IS CLAIMED IS:

A phosphate glassification treatment method for radioactive boric acid liquid waste, wherein phosphate glass is created by adding, to concentrated boric acid liquid waste made by concentrating radioactive boric acid created at a nuclear power facility, phosphoric acid and an alkali metal or an alkali metal salt of phosphoric acid so as to achieve a composition in the range bounded by the lines indicating B<sub>2</sub>O<sub>3</sub> 20 mol% and 40 mol% and the lines indicating X<sub>2</sub>O 20 mol% and 40 mol% in the glass composition range diagram shown in FIG. 1 (in the diagram, X represents an alkali metal, and XO<sub>2</sub> represents the molarity in a combination of one or more of Li<sub>2</sub>O, Na<sub>2</sub>O, and K<sub>2</sub>O), and kneading and heating this to remove water and remove the water of crystallization, producing a calcinate consisting mainly of a boron phosphate compound; the calcinate is heated to 600°C or higher to create a phosphate glass.

- 3. DETAILED DESCRIPTION OF THE INVENTION

### Industrial Field of Application

The present invention relates to a phosphate glassification treatment method, for radioactive boric acid liquid waste, which converts the radioactive boric acid liquid waste created by a nuclear power facility, especially a pressurized water nuclear reactor (hereinafter "PWR"), into a stable solid.

## Prior Art

In conventional practice, the radioactive boric acid liquid waste created at a PWR power generating station, etc. is vaporized and concentrated, and then sent to an asphalt solidification treatment apparatus, and while it is being mixed with asphalt the remaining moisture component is vaporized and removed at high temperature (120°C or lower), and finally is converted into a solid form kneaded into asphalt, and is put into drums and stored.

## Problems the Invention Is to Solve

However, the abovementioned asphalt solidification method has the following sort of problems that are defects caused by the essential nature of asphalt, and is not suitable for the final (permanent) storage and treatment methods (for example, disposal at sea, underground storage, throwing away) envisioned in the near future.

- (1) The solidified asphalt is broken down by heat and light (radiation) and generates gas.
- (2) The solidified asphalt is combustible and oxidizes.
- (3) The solidified asphalt is deficient in mechanical strength, and is thermoplastic.
- (4) The solidified asphalt is not dense enough.
- (5) The solidified asphalt is deficient in volume decrease. For example, a PWR standard unit plant (1200 MWe  $\times$  2 reactors) creates 150 tons of 12% radioactive concentrated boric acid liquid waste per year. The solidified asphalt from this would fill 200 200-liter drums.

Because of this sort of background, development and research has been proceeding on methods of converting liquid waste into an inorganic solid that are suitable for final disposal methods and achieve even greater volume reduction.

The present invention is one that was created in order to solve the sort of problems described above.

## Means for Solving the Problems

A radioactive boric acid liquid waste treatment method for dealing with the sort of current situation and problems described above basically needs to have the following sort of characteristics.

- (1) It is a process that removes the remaining moisture component from concentrated boric acid liquid waste, and in addition promotes a reaction to remove the water of crystallization ( $2 \text{H}_3\text{BO}_3 \rightarrow \text{B}_2\text{O}_3 + 3 \text{H}_2\text{O}$ ) with little possibility of scattering radioactive material (radioactivity management).
- (2) This process is relatively simple, and has low operating costs from the perspective of the required power, etc. (operating maintenance, economy).
- (3) Boric acid is incorporated into its matrix, resulting in a solid that is chemically and physically stable (compatibility with waste standards).
- (4) As much boric acid as possible is incorporated into the solid without impairing the stability of the solid (volume reduction).

The present invention is one that was created based on such concepts. More specifically, it prepares the initial material by adding, to concentrated boric acid liquid waste, phosphoric acid and an alkali metal or an alkali metal salt of phosphoric acid so as to achieve a composition in the slanted-line range bounded by the lines indicating  $\text{B}_2\text{O}_3$  20 mol% and 40 mol% and the lines indicating  $\text{X}_2\text{O}$  20 mol% [and 40 mol%]<sup>1</sup> in the FIG. 1 glass composition range, thereby forming a molten salt that can be kept at in a liquid phase state to high temperature, and therefore

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<sup>1</sup> This phrase omitted in original.

proceeding with a moderate condensation-dewatering reaction that causes almost no scattering of radioactive material. However, in FIG. 1, X represents an alkali metal, and  $XO_2$  represents the total molarity in a combination of one or more of  $Li_2O$ ,  $Na_2O$ , and  $K_2O$ . As the reaction progresses, the molten liquid becomes more viscous, and ultimately creates a calcinate (powder) consisting of a boron phosphate compound containing almost no moisture.

In addition, the calcinate obtained in this manner is heated to  $600^\circ C$  or higher to form a homogeneous phosphate glass. The phosphate glass created in this manner has  $-\{O-P-O-P-O\}-$  as its glass network structure, so the percentage of boric acid contained is high, and it has properties that are stable chemically and mechanically.

### Operation

A molten salt that can be kept at in a liquid phase state to high temperature is formed by adding an alkali metal salt of phosphoric acid to concentrated boric acid liquid waste, and while this liquid phase state is maintained, vaporization of the remaining moisture component and a condensation/dewatering reaction proceeds, forming larger molecules (making the molten liquid more viscous) and ultimately creating a powder calcinate consisting of a boron phosphate compound. The calcinate that is formed can be made into a stable and homogeneous phosphate glass using an electric furnace, etc.

### Embodiments

Below, embodiments of the present invention shall be described in detail.

In the present invention, alkali metal salts include  $Li_2CO_3$ ,  $Na_2CO_3$ ,  $K_2CO_3$ , etc., which are carbonates of alkali metals;  $LiNO_3$ ,  $NaNO_3$ ,  $KNO_3$ , etc., which are nitrates of alkali metals; and  $Li_2SO_4$ ,  $Na_2SO_4$ ,  $K_2SO_4$ , etc., which are sulfates of alkali metals. Alkali metal salts of phosphoric acid include  $LiH_2PO_4$ ,  $NaH_2PO_4$ ,  $KH_2PO_4$ , etc., which are primary phosphates of alkali metals, and  $Li_2HPO_4$ ,  $Na_2HPO_4$ ,  $K_2HPO_4$ , etc., which are secondary phosphates of alkali metals.

The region of the slanted lines shown in FIG. 1 was determined through consideration of the following points.

- (1) Making the melting-glassification temperature as low as possible.
- (2) Going in the direction approaching  $B_2O_3$  in FIG. 1 so as to contain as much as possible of the boric acid component.
- (3) Making the range as far as possible from the vertices of  $X_2O$ ,  $P_2O_5$ , and  $B_2O_3$  so as to be chemically stable. Water resistance in particular becomes dramatically worse near the  $X_2O$  vertex.

In all of the following embodiments, the starting material was prepared so as to produce components per the abovementioned slanted lines.

Embodiment 1: When B/P (atomic ratio) = 1/1 using primary sodium phosphate

A sample was made by adding 303g of primary sodium phosphate ( $NaH_2PO_4 \cdot 2 H_2O$ ) to 1 kg of concentrated boric acid liquid (12 wt% as  $H_3BO_3$ ). This amount of primary sodium phosphate was determined so that the atomic ratio of B/P would be 1/1 in the calcinate composition after dewatering and the end of calcination.

This sample was placed in a suitable container, and slowly heated while stirring and mixing it; excess moisture other than water of crystallization contained in the sample was

vaporized and removed at 80~100°C. As a result the sample became a molten salt that was rather viscous and highly homogeneous.

While continuing to stir the sample, it was additionally heated and its temperature was gradually raised from 100°C to 200°C. Repeated condensation and dewatering reactions were performed while keeping the sample in a liquid phase state; it passed through a gel-like high-viscosity state and became a powder.

Finally it was heated to 300~400°C and became a complete calcinate (granules). About 265g of calcinate was obtained, consisting of two compounds ( $\text{Na}_2\text{BPO}_5$ ,  $\text{BPO}_4$ ). The reaction, expressed as a chemical reaction, was  $2 \text{H}_3\text{BO}_3 + 2 \text{NaH}_2\text{PO}_4 \rightarrow \text{Na}_2\text{BPO}_5 + \text{BPO}_4 + 4 \text{H}_2\text{O}^\uparrow$ .

The calcinate thus created was placed in a magnetic crucible, and heated and melted in an electric furnace at 700°C to make a glass solid. The resulting glass was a phosphate glass whose basic unit was  $-\{\text{O}-\text{P}-\text{O}-\text{P}-\text{O}\}-$ , and its density was about 2.3. It was possible to reduce the volume to about 1/9 of the original volume of concentrated boric acid liquid.

Embodiment 2: When B/P (atomic ratio) = 1/2 using primary sodium phosphate

A sample was made by adding 606g of primary sodium phosphate ( $\text{NaH}_2\text{PO}_4 \cdot 2 \text{H}_2\text{O}$ ) to 1 kg of concentrated boric acid liquid (12 wt% as  $\text{H}_3\text{BO}_3$ ), as in Embodiment 1.

About 450g of glass solid was obtained from this sample through exactly the same operation as in Embodiment 1. The density of the glass was about 2.2, and it was possible to reduce the volume to about 1/5 of the original concentrated boric acid liquid.

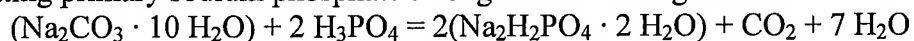
Embodiment 3: When B/P (atomic ratio) = 2/1 using primary sodium phosphate

A sample was made by adding 152g of primary sodium phosphate ( $\text{NaH}_2\text{PO}_4 \cdot 2 \text{H}_2\text{O}$ ) to 1 kg of concentrated boric acid liquid (12 wt% as  $\text{H}_3\text{BO}_3$ ), as in Embodiments 1 and 2.

About 167g of glass solid was obtained from this sample through exactly the same operation as in Embodiments 1 and 2. The density of the glass was about 2.3×, and it was possible to reduce the volume to about 1/10 of the original concentrated boric acid liquid.

Embodiment 4: When using phosphoric acid and sodium carbonate instead of primary sodium phosphate

A sample was made by adding 280g of sodium carbonate ( $\text{Na}_2\text{CO}_3 \cdot 10 \text{H}_2\text{O}$ ) and 190g of phosphoric acid ( $\text{H}_3\text{PO}_4$ ) to 1 kg of concentrated boric acid liquid (12 wt% as  $\text{H}_3\text{BO}_3$ ). These amounts of sodium carbonate and phosphoric acid were determined based on the stoichiometric ratio for creating primary sodium phosphate through the following reaction.



About 265g of phosphate glass solid was finally obtained from this sample through exactly the same operation as in Embodiment 1. The characteristics of the resulting solid were the same as those obtained in Embodiment 1.

Embodiment 5: When using primary potassium phosphate instead of primary sodium phosphate

A sample was made by adding 270g of primary potassium phosphate ( $\text{KH}_2\text{PO}_4$ ) to 1 kg of concentrated boric acid liquid (12 wt% as  $\text{H}_3\text{BO}_3$ ). About 270g of phosphate glass solid was obtained from this sample through exactly the same operation as in Embodiment 1. The characteristics of the resulting solid were similar to those obtained in Embodiment 1.

### Effect of the Invention

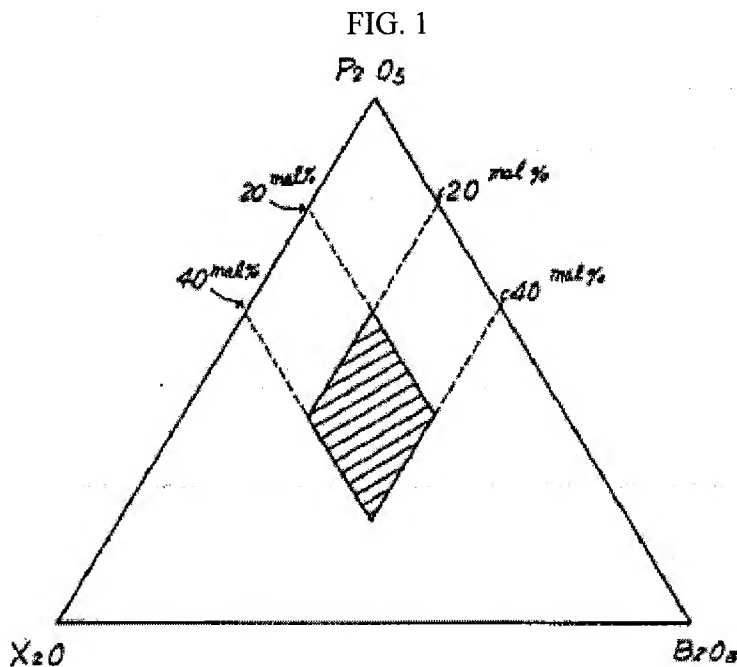
The phosphate glassification treatment method for radioactive boric acid of the present invention as described in detail above achieves the following sorts of effects.

- (1) The condensation-dewatering reaction takes place while maintaining a liquid phase state, so there is very little creation of radioactive fume dust, etc.
- (2) The operation is carried out continuously, and the process is simple.
- (3) Phosphoric acid is low in price, and if it is used, calcinations and glassification can be done at a relatively low temperature, so it is very economical.
- (4) The resulting glass solid has high strength and high density ( $2.2\sim 2.3\text{ g/cm}^3$ ), and has excellent chemical stability.
- (5) The resulting glass solid has a high boric acid content ( $15\sim 30\text{ wt\% as B}_2\text{O}_3$ ), and is excellent in volume reduction.

Accordingly, compared to the asphalt solidification method it is possible to reduce the amount of drums generated per year by about 50% or more. (Based on the 150 tons of 12% concentrated boric acid liquid waste per year created by a PWR standard unit plant with 1200 MWe  $\times$  2 reactors, the glass solid produced by the present method would be about 100 200-liter drums.)

#### 4. BRIEF DESCRIPTION OF THE DRAWINGS


FIG. 1 is a composition diagram showing the composition range of phosphate glass.





## **CERTIFICATE OF ACCURACY**

This is to certify that the attached document, *Japanese Patent JP63015200A*, has been translated from Japanese into English by staff members of LANGUAGE MATTERS familiar with the Japanese and English languages, and is to the best of our knowledge, ability and belief a true and accurate translation.



For LANGUAGE MATTERS

Sworn to and subscribed before me  
this 21<sup>st</sup> day of August, 2009



**NOTARY**

JUDITH E. MORRIS  
Notary Public  
State of Colorado  
My Commission Expires 07/03/2012

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審査請求 未請求 発明の数 1 (全4頁)

⑭ 発明の名称 放射性ほう酸廃液のりん酸塩ガラス化処理法

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## 明 細 書

## 1. 発明の名称

放射性ほう酸廃液のりん酸塩  
ガラス化処理法

## 2. 特許請求の範囲

原子力施設で発生する放射性ほう酸廃液を濃縮したほう酸濃縮廃液に、処理対象とするほう酸量に対応したりん酸とアルカリ金属もしくはりん酸のアルカリ金属塩を第1図に示すガラスの組成範囲図〔図中Xはアルカリ金属を表し、 $X_2O$ は $Li_2O$ ,  $Na_2O$ ,  $K_2O$ の1種または2種以上の組合せにおけるモル数を表す〕の $B_2O_3$ : 20mol%, 40mol%, を示すラインと $X_2O$ : 20mol%, 40mol%を示すラインで囲まれる領域の組成になるように添加し、これを混練ー加熱することにより、脱水と脱結晶水を図り、りん酸ほう素化合物を主体とする仮焼体とし、該仮焼体を600℃以上に加熱することにより、りん酸塩ガラスを生成せしめることを特徴とする放射性ほう酸廃液のりん酸塩ガラス化処理法。

## 3. 発明の詳細な説明

(産業上の利用分野)

本発明は加圧水型原子炉(以下PWRと称す)をはじめとする原子力施設で発生する放射性ほう酸廃液を安定固化体化する放射性ほう酸廃液のりん酸塩ガラス化処理法に関するものである。

(従来技術)

従来、PWR発電所等で発生する放射性ほう酸廃液は蒸発濃縮された後、アスファルト固化処理装置に送られ、ここでアスファルトと混合されながらさらに高温(120℃以下)で残存水分が蒸発除去され、最終的にはアスファルトに混練された形の固化体に転換されドラム缶詰されて貯蔵されている。

〔発明が解決しようとする問題点〕

しかし、上記アスファルト固化法は、アスファルトの本質に起因する欠点として次のような問題があり、近い将来に想定されている最終(永久)貯蔵・処分法(たとえば海中投棄、地中貯蔵、投棄など)には適合しない。

- ① アスファルト固化体は熱や光（放射線）で分解しガスを発生する。
- ② アスファルト固化体は可燃性で酸化される。
- ③ アスファルト固化体は機械的強度に劣り、熱可塑性である。
- ④ アスファルト固化体は十分な密度がない。
- ⑤ アスファルト固化体は減容性に劣る。

例えばPWR標準ユニットプラント（1200MWe×2基）からは年間150トンの12%放射性ほう酸濃縮廃液が発生し、これからアスファルト固化体は200ℓドラム缶で200本分のものとなる。

このような背景があるため最終処分法に適合し、かつよりすぐれた減容化が得られる無機固化体へ転換処理する方法の開発・研究が進められている。

本発明は、上記のような問題点を解決するためになされたものである。

〔問題点を解決するための手段〕

前述のような現状・問題点に対処するための放射性ほう酸廃液処理法としては基本的に次のよう

るよう添加して原料を調整し、高温まで液相状態を維持できる熔融塩とし、そこで放射性物質飛散がほとんど起こらない穏やかな縮合－脱水反応を進行させる。但し、第1図においてXはアルカリ金属を表し、 $X_2O$ は $Li_2O$ 、 $Na_2O$ 、 $K_2O$ の1種または2種以上の組合せにおける合計のモル数を表す。こうして反応の進行と共に融液の高粘性化が進み、最後に水分の全く含まないりん酸ほう素化合物からなる仮焼体（粉末）ができる。

さらに、こうして得られた仮焼体を600℃以上に加熱することにより均質なりん酸塩ガラスができる。こうしてできるりん酸塩ガラスとは $-[O-P(=O)(OH)_2-O-P(=O)(OH)_2-O]-$ をそのガラス網目構造とするもので、ほう酸含有率が大きく、しかも化学的・物理的に安定な性質を有する。

〔作用〕

ほう酸濃縮廃液にりん酸のアルカリ金属塩を加えることにより、高温まで液相状態を持続する熔融塩となり、その液相状態を持続させながら残存水分の蒸発－縮合脱水反応を進めると、高分子化

な特徴を有することが必要である。

- ① ほう酸濃縮廃液から残存水分を除き、さらに脱結晶水反応（ $2H_3BO_3 \rightarrow B_2O_3 + 3H_2O$ ）を進めるプロセスは放射性物質飛散の可能性の小さいものであること（放射線管理）。
- ② そのプロセスは比較的単純で、かつ必要動力等の面からみて運転経費が少ないこと（運転保守性・経済性）。
- ③ ほう酸をそのマトリックス内に取り込み、化学的・物理的に安定な固化体が得られること（廃棄物基準適合性）。
- ④ 固化体の安定性を損なわない範囲でなるべく多くのほう酸を固体内に取り込むこと（減容性）

本発明はこのような考え方に基づいてなされたものである。すなわち、ほう酸濃縮廃液にりん酸とアルカリ金属もしくはりん酸のアルカリ金属塩を第1図のガラスの組成範囲図の $B_2O_3$ ：20mol%、40mol%を示すラインと $X_2O$ ：20mol%を示すラインで囲まれる斜線の領域の組成にな

（融液の高粘性化）し最終的にりん酸ほう素化合物からなる粉末仮焼体となり、できた仮焼体は電気炉等により比較的低温で安定で均質なりん酸塩ガラスとなる。

〔実施例〕

以下、本発明の実施例を詳細に説明する。

本発明において、アルカリ金属塩とは、アルカリ金属の炭酸塩である $Li_2CO_3$ 、 $Na_2CO_3$ 、 $K_2CO_3$ 等、アルカリ金属の硝酸塩である $LiNO_3$ 、 $NaNO_3$ 、 $KNO_3$ 等、アルカリ金属の硫酸塩である $Li_2SO_4$ 、 $Na_2SO_4$ 、 $K_2SO_4$ 等であり、りん酸のアルカリ金属塩とは、アルカリ金属の第1りん酸塩である $LiH_2PO_4$ 、 $NaH_2PO_4$ 、 $KH_2PO_4$ 等、アルカリ金属の第2りん酸塩である $LiHPO_4$ 、 $NaHPO_4$ 、 $KHPO_4$ 等である。

第1図の斜線の範囲は下記の点を勘案して決定した。

- ① 熔融－ガラス化温度がなるべく低いこと。
- ② ほう酸成分をなるべく多く含有できるように第1図で $B_2O_3$ に近づく方向にする。

③ 化学的に安定になるように  $X_2O$ ,  $P_2O_5$ ,  $B_2O_3$  の頂点からなるべく遠い範囲にする。特に  $X_2O$  の頂点に近いと極端に耐水性が悪くなる。

以下の実施例は総て上記斜線の成分となるよう原料を調整した。

実施例1 …りん酸第1ナトリウムを用い  $B/P = 1/1$  (原子比) の場合。

濃縮ほう酸水 (12 Wt% as  $H_3BO_3$ ) 1 kg に 303 g のりん酸第1ナトリウム ( $NaH_2PO_4 \cdot 2H_2O$ ) を加え試料とする。このりん酸第1ナトリウムの量は脱水・仮焼終了後の仮焼体組成において  $B/P$  の原子比が  $1/1$  になるようにきめられた。

この試料を適当な容器に入れ、攪拌・混合しながらゆっくりと加熱し、80～100℃で試料中に含まれる結晶水以外の余剰水分を蒸発・除去する。こうして試料はかなり粘性の高い均質な溶融塩となる。

試料の攪拌を続けながら、さらに100℃から200℃へと徐々に加熱昇温する。試料は液相状態を維持しつつ縮合・脱水反応を繰り返し水アメ

2.2であり、もとの濃縮ほう酸水に比較して約1/5に減容化できた。

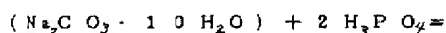
実施例3 …りん酸第1ナトリウムを用い  $B/P = 2/1$  (原子比) の場合。

実施例1及び2と同様に濃縮ほう酸水 (12 Wt% as  $H_3BO_3$ ) 1 kg に 152 g のりん酸第1ナトリウム ( $NaH_2PO_4 \cdot 2H_2O$ ) を加え試料とする。

この試料を実施例1及び2と全く同じ操作により約187gのガラス固化体を得た。ガラスの密度は約2.3倍でありもとの濃縮ほう酸水に比較して約1/10に減容化できた。

実施例4 …りん酸第1ナトリウムを代えてりん酸と炭酸ナトリウムを用いた場合。

濃縮ほう酸水 (12 Wt% as  $H_3BO_3$ ) 1 kg に 280 g の炭酸ナトリウム ( $Na_2CO_3 \cdot 10H_2O$ ) 及び 190 g のりん酸 ( $H_3PO_4$ ) を加え試料とする。この炭酸ナトリウム及びりん酸の量は次の反応によりりん酸第1ナトリウムを生成する化学量論比に基づいて決めた。



状の高粘性状態を経てついに粉末化する。

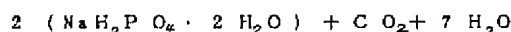
最後に300～400℃に加熱し完全な仮焼体 (粒状) とする。こうして約265gの仮焼体を得られこれは二種の化合物 ( $Na_2B_4P_2O_{12}$ ,  $B_2P_2O_7$ ) からなっており、その反応を化学反応式で示すと、  
 $2H_3BO_3 + 2NaH_2PO_4 \rightarrow Na_2B_4P_2O_{12} + B_2P_2O_7 + 4H_2O$  となる。

こうしてできた仮焼体を磁性ルツボに入れ、電気炉で700℃に加熱・溶融し、ガラス固化体を得る。得られたガラスは  $-[O-\overset{\text{P}}{\underset{\text{O}}{\text{B}}}-O-\overset{\text{P}}{\underset{\text{O}}{\text{B}}}-O]-$  を基本単位とするりん酸塩ガラスであり、密度は約2.3であった。こうしてもとの濃縮ほう酸水の容積に比べ約1/9に減容化することができた。

実施例2 …りん酸第1ナトリウムを用い  $B/P = 1/2$  (原子比) の場合。

実施例1と同様に濃縮ほう酸水 (12 Wt% as  $H_3BO_3$ ) 1 kg に 606 g のりん酸第1ナトリウム ( $NaH_2PO_4 \cdot 2H_2O$ ) を加え試料とする。

この試料を実施例1と全く同じ操作により約450gのガラス固化体を得た。ガラスの密度は約



この試料を実施例1と全く同様の操作により最終的に約265gのりん酸塩ガラス固化体を得た。得られた固化体の特徴は実施例1で得られたものと同じであった。

実施例5 …りん酸第1ナトリウムに代えてりん酸第1カリウムを用いた場合。

濃縮ほう酸水 (12 Wt% as  $H_3BO_3$ ) 1 kg に 270 g のりん酸第1カリウム ( $KH_2PO_4$ ) を加え試料とした。この試料を実施例1と全く同様の操作により約270gのりん酸塩ガラス固化体を得た。得られた固化体の特徴は実施例1で得られたものと類似したものであった。

〔発明の効果〕

以上詳細に説明した本発明の放射性ほう酸廃液のりん酸塩ガラス化処理法によれば、下記の如き効果を奏する。

① 液相状態を維持しながら縮合・脱水反応が進むため放射性ヒューム・グスト等の発生がきわめて少ない。

② 操作が連続的に行え、またプロセスが単純である。

③ りん酸塩は価格が低廉であり、またこれを用いれば比較的低温で仮焼、ガラス化ができるため経済性にすぐれている。

④ 得られるガラス固化体は高強度・高密度 ( $2.2 \sim 2.3 \text{ g/cm}^3$ ) で化学的安定性にもすぐれている。

⑤ 得られるガラス固化体はほう酸含有率が大きく ( $15 \sim 30 \text{ wt\% as B}_2\text{O}_3$ )、溶解性に優れている。

そのため、アスファルト固化体法にくらべて発生する年間ドラム缶量は約50%以上の低減化が可能である。(PWR標準ユニットプラント1200MW $\times$ 2基で発生する150トンの12%ほう酸濃縮廃液から本方法によるガラス固化体として200 $\ell$ ドラム缶で約100本分ができる)

#### 4. 図面の簡単な説明

第1図はりん酸塩のガラスの組成範囲を示す組成図である。

